

New aliphatic–aromatic tetraphenylphthalic-based diimides: Thermal, optical and electrical study

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ABSTRACT

The thermal and opto-electrical properties of new thermotropic compounds based on tetraphenylphthalic-based imides have been explored. Energy band gap (E_g^{opt}) was calculated from absorption spectrum and absorption coefficient α . Current–voltage (I – V) measurements were performed on ITO/compound/Al, ITO/compound:PCBM/Al and ITO/PEDOT:PSS/compound:PCBM/Al devices before and after irradiation with light (under illumination 1000 W/m²). Compounds were blended with [6,6]-phenyl C₆₁ butyric acid methyl ester (PCBM) in the weight ratio 1:1 and additionally tested using various AFM techniques. The mesogenic properties were characterized by POM and DSC. Both compounds exhibited liquid crystalline properties. Preliminary photovoltaic experiments were carried out for two kinds of bulk heterojunction devices (BHJ) and compared with reference device. For the first time, to the best of our knowledge, tetraphenylphthalic-based imides with liquid crystalline properties were examined and described in the article.

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1. Introduction

During past decade, considerable attention has been focused on new materials applicable in organic electronics devices such as light emitting diodes (OLEDs), solar cells, and field effect transistors (FETs) [1–3]. For some of these materials self-organization might supply an efficient path for charge transporting [4]. Low and high molecular weight liquid crystals (LCs) are particularly interesting owing to their ability to form large aligned domains, which enable the formation of well-ordered thin films [2]. Organic semiconductors can function either as p-channel or n-channel charge carrier. Although impressive progress was done in developing electron-rich character (p-type) organic materials, the pursuit for n-type semiconductors, capable of electron transport, still lagged behind that of p-type systems [1]. A lot of effort have been devoted to synthesizes of n-type organic semiconductors with high electron mobilities, good environmental stability, and good processability [1]. To allow for efficient electron injection from

common metal electrodes, a critical threshold electron affinity of 3.0 eV is required for n-type semiconductor materials [5].

Among many designed and synthesized organic materials, compounds with imide groups are particularly interesting. It is expected that introduction of electron-withdrawing dicarboxylic imide units into compounds may increase the electron affinity in conjugation with thermal, air and chemical stability. Since the report of naphthalenetetracarboxylic diimides as air stable n-channel materials, a large number of either naphthalene or perylene tetracarboxylic diimides has been synthesized and investigated [6–11]. Such imides contain six-membered imide rings.

Other kinds of imides are compounds with five-membered imide rings, that are, tetracarboxylic diimides. It is quite surprising that they are only occasionally investigated as semiconductors. To the best of our knowledge, only three articles described such investigations [2,12,13]. Mark et al. reported a new electron-deficient semiconductors based on the anthracenedicarboxyimide [12]. Pyromellitic diimide derivatives were investigated as materials for organic field effect transistors by Katz et al. [2] Dingemans et al. studied diimides from 3,3',4,4'-biphenyltetracarboxylic dianhydride (BPDA) and meta-substituted aryl-ether amines, which exhibited excellent electron injection properties [13].

On the other hand aromatic imides group are almost planar, rigid, polar and thermostable, and thus should be favorable components of liquid crystalline (LC) polymers regardless of whether

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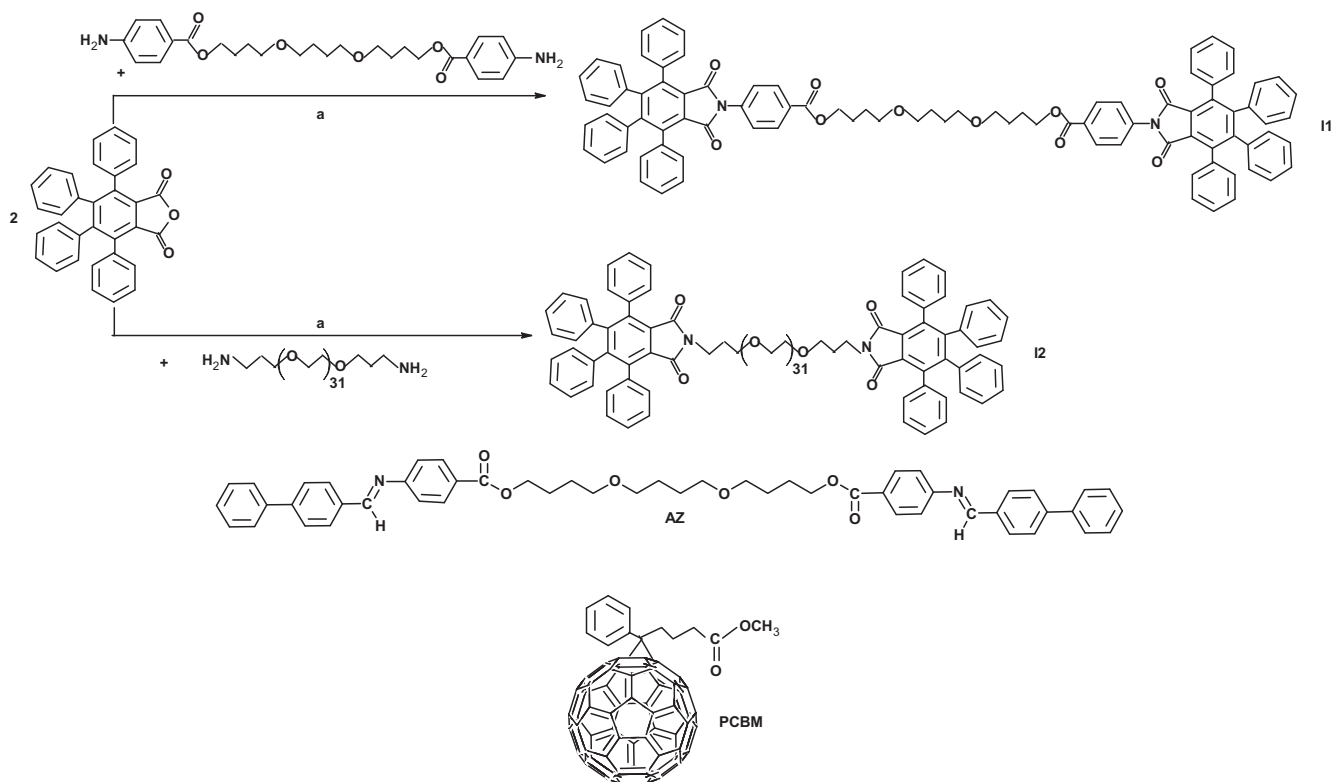


Fig. 1. Synthetic route and chemical structure of diimides along with chemical structure of azomethine **AZ**. a: NMP, 180 °C, 9 h. Chemical structure of [6,6]-phenyl C₆₁ butyric acid methyl ester (PCBM).

thermotropic or lyotropic character is taken into consideration [14]. A variety of liquid crystalline five-membered imides have been studied so far including both imides and diimides [13,15–18]. It was found that in the case of low molecular weight compounds an phthalimide unit is not an efficient mesogen [15,16]. However, N-substituted cholesteryl esters of the 4-carboxyphthal-diimides formed a chiral nematic and smectic A or C phases [15,18], a nematic phase showed biphenyl-4,4'-diyl bis(N-hexylphthalimide-4-carboxylate) [16] as well as phthalimides with ester and oxyethylene groups revealed the smectic behavior [17].

It seems to be promising to obtain five-membered imides with liquid crystalline properties as potential organic semiconductors. To the best of our knowledge, only in one article such idea was utilized [13]. Dingemans et al. obtained ether-imides by terminating various dianhydrides with flexible aryl-ether tails of different chain lengths, which exhibited the classic calamitic “tail-core-tail” structure [13].

Both low and high molecular weight imides are well known high performance materials, whose applications range from aerospace to microelectronics, (opto)electronics and photonics [19]. The main reason of the interest in imides originates from their electron acceptor properties. It should be emphasized that aromatic imide rings, with electron deficient cores, have demonstrated a great potential as n-type (electron-transporting) semiconductors to which much less attention has been paid in comparison with the p-type organic semiconductors [20]. On the other hand aromatic imide groups are almost planar, rigid, polar and thermostable, and thus should be favorable components of liquid crystalline compounds as good mesogens [14,15]. Moreover, for the service life of photovoltaic cells, organic compounds should be stable upon being subjected to thermal, chemical and photo-physical stimuli. It was found that typical conjugated polymers are usually not stable enough [21]. Therefore, the design and preparation of photoactive organic compounds that not only have an

extended absorption edge, to match the solar terrestrial radiation, and a high carrier mobility, but also good thermal and chemical stabilities, to improve the lifetime of OPV devices, is regarded as the main challenge for organic semiconductors [22]. The introduction of imide rings preserves the thermal and chemical stability of compounds. Inspired by the remarks described above we have undertaken a preparation and investigation of new LC diimides.

On the basis of these promising results we designed new LC imides with “core-tail-core” structure, presented in this paper, which can constitute perspectives for family of materials for optoelectronic applications. We have chosen for preparation of such diimides anhydride with extended aromatic sphere, that is, 3,4,5,6-tetraphenylphthalic acid anhydride and two diamines: 4,4'-(butane-1,4-diylbis(oxy))bis(butane-4,1-diyl) bis(4-amino-benzoate) (**PBBA470**) and poly(ethyleneglycol)-bis-(3-aminopropylether) (**PEG1500**).

Recently, we described the thermal and opto-electrical properties (absorption, photoluminescence, thermoluminescence) of symmetrical azomethine based on the **PBBA470** [23]. This azomethine exhibited SmA and SmB mesophases what was confirmed by DSC, POM and X-ray diffraction.

This paper is devoted to synthesis of two thermotropic diimides with or without ester groups in the aliphatic part and their thermal (DSC, POM), optical (UV–vis), morphological (AFM) and electrical (*I*–*V*) characterisations summary by preliminary photovoltaic experiments.

2. Results and discussion

2.1. Synthesis and characterisation

Two novel LC diimides in which extended aromatic sphere imide groups were linked by flexible spacer containing aryl-ester,

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